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Generalized Test Plan for the Vitrification of Simulated High-Level-Waste Calcine in the Idaho National Laboratory's Bench-Scale Cold Crucible Induction Melter

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Summary

This Preliminary Idaho National Laboratory (INL) Test Plan outlines the chronological steps required to initially evaluate the validity of vitrifying INL surrogate (cold) High –Level –Waste (HLW) solid particulate calcine in INL's Cold Crucible Induction Melter (CCIM). Its documentation and publication satisfies interim milestone WP-413-INL-01 of the DOE – EM (via the Office of River Protection) sponsored work package, WP 4.1.3, entitled "Next Generation Induction-Heated and Advanced Joule-Heated Melter Bench Scale Testing" A more detailed test plan will be issued at least 2 weeks before the actual CCIM/calcine test once all glass formulation studies and CCIM modifications are completed.

The primary goal of the proposed CCIM testing is to initiate efforts to identify an efficient and effective back – up and risk adverse technology for treating the actual HLW calcine stored at the INL. The calcine's treatment must be completed by 2035 as dictated by a State of Idaho Consent Order. A final report on this surrogate /calcine test in the CCIM will be issued in May 2012-pending next fiscal year funding.

In particular the plan provides; 1) distinct test objectives, 2) a description of the purpose and scope of planned university contracted pre-screening tests required to optimize the CCIM glass/surrogate calcine formulation,3) a listing of necessary equipment modifications and corresponding work control document changes necessary to feed a solid particulate to the CCIM,4) a description of the class of calcine that will be represented by the surrogate, and 5) a tentative tabulation of the anticipated CCIM testing conditions, testing parameters, sampling requirements and analytical tests. Key FY -11 milestones associated with this CCIM testing effort are also provided.

The CCIM test run is scheduled to be conducted in February of 2012 and will involve testing with a surrogate HLW calcine representative of only 13% of the 4,000 m^3 of "hot" calcine residing in 6 INL Bin Sets. The remaining classes of calcine will have to be eventually tested in the CCIM if an operational scale CCIM is to be a feasible option for the actual INL HLW calcine. This remaining calcine's make-up is HLW containing relatively high concentrations of zirconium and aluminum, representative of the cladding material of the reprocessed fuel that generated the calcine. A separate study to define the CCIM testing needs of these other calcine classifications with an emphasis placed on the formation of glass-ceramics (as opposed to only glass) is currently being prepared under a separate work package (WP-0) and will be provided as a milestone report at the end of this fiscal year.

Contents

ACRONYMS

BEA	Battelle Energy Alliance
BSG	Borosilicate Glass
CCIM	Cold Crucible Induction Melting
CO/CR	Carry over/Continuing resolution
DOE	Department of Energy
EM	US DOE Department of Environmental Management
FY	Fiscal Year
HIP	Hot Isostatic Press
HLW	High-Level- Waste
ICP	Idaho Clean-up Project
IEDF	INL Engineering Demonstration Facility
INL	Idaho National Laboratory
IWTU	Idaho Waste Treatment Unit
JHM	Joule Heated Melter
LI	Laboratory Instruction (BEA Work Control Document for CCIM Testing)
NWCF	New Waste Calcining Facility
PCT	Product Consistency Test
RCRA	Research Conservation and Recovery Act
ROD	Record of Decision
SRS	Savannah River Site
SOW	Scope of Work
TCLP	Toxic Characteristic Leaching Procedure
VHT	Vapor Hydration Test
WP	Work Package

Generalized Test Plan for the Vitrification of Simulated High –Level -Waste Calcine in the Idaho National Laboratory's Bench -Scale Cold Crucible Induction Melter

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1. Background

The Department of Energy's (DOEs) Idaho National Laboratory (INL) - through its clean-up contractor attached to the Idaho Clean-up Project (ICP) - is required under a State of Idaho agreement to remove and treat, for eventual disposal, over 4,000 m^3 of solid radioactive High- Level- Waste (HLW) calcine by 2035. The HLW calcine was generated as a result of reprocessing Defense and unique nuclear reactor fuels for over 40 years. Since the reprocessing method was an aqueous technique, the calcine was generated by solidifying the liquid raffinate in a de-nitrating fluidized bed to a particulate with an average diameter of ~3 mm. Even though the calcine step reduced the HLW raffinate volume by over eight fold, the calcine, which is currently stored in 6 different vaulted bin sets, is not acceptable for direct disposal in any future HLW/spent fuel repository. As such, the calcine must be properly immobilized to contain its fission products, long-term alpha emitting radionuclides (unrecovered actinides and transuranics) and hazardous RCRA metals.

To accomplish the calcine treatment objectives, the Idaho Clean-up Project contractor, CWI, has chosen to immobilize the calcine in a glass-ceramic via the use of a Hot-Isostatic-Press (HIP); a treatment selection formally documented in a 2010 Record of Decision (ROD). The HIP process is currently under both development and design and present plans call for adding HIPing via modifications to the Idaho Waste Treatment Unit (IWTU); a facility recently constructed for treating liquid radioactive tank waste.
Even through the HIP process may prove suitable for the calcine as specified in the ROD and validated in a number of past Value Engineering sessions; the Department of Energy is evaluating back-up treatment methods for the calcine as a result of the technical, schedule, and cost risk associated with the HIPing process. Consequently DOE-HQ has requested DOE-ID to make INL's bench-scale cold-crucible induction melter (CCIM) available for investigating its viability as a process alternate to calcine treatment.

With that back-drop, this document provides a generalized test plan for initial vitrification testing of simulated non –radioactive calcine in INLs bench scale CCIM. After an introduction section, test objectives are provided, followed by tentative test run conditions, test parameters and proposed sampling and analysis requirements. This plan also provides descriptions of auxiliary pre- test run activities that must be completed to ensure successful completion of the CCIM/calcine test. These activities include CCIM bench scale equipment modifications and all associated work control documents, initiation of small scale pre-screening tests to be performed by a university (known for their research and development in radioactive waste immobilization technologies and related material science) and calcine simulant preparation and procurement of the first type of calcine simulant to be tested. A more detailed run plan for actual test operations will be available at a later date prior to the actual scheduled testing in FY-12. This final test plan will incorporate all specific details of test conditions and parameters to ensure objectives are met and identify future calcine/CCIM testing. This final test plan will also define the complete expected contents of the future results report as well as contain requirements for data quality and quality assurance

2. Introduction

The INL's bench-scale Cold Crucible Induction melter (CCIM) is located in Idaho Falls, Idaho at the DOE's owned Idaho Engineering Development Facility- IEDF (Pilot-Plant test bay number 1) and will be the location of the test described in this plan. Details of the bench scale CCIM and its unique technology can be found in various references (1 and 2) and are not being provided here. For general information, a simplified schematic of the major CCIM systems is shown in Figure 1 below and a photo of the 10-cm diameter CCIM crucible (showing induction coils) during the draining of melted glass/waste is shown in Figure 2. Both figures highlight the major design features that provide improvements over the conventional Joule Heated Melters currently in wide use for HLW. These features include the induction coils, which eliminate in-bed electrodes subject to high corrosion; and the vertical cooling tubes that make up the crucible wall, which in –turn create a sold glass wall that negates the need for corrosive refractory.

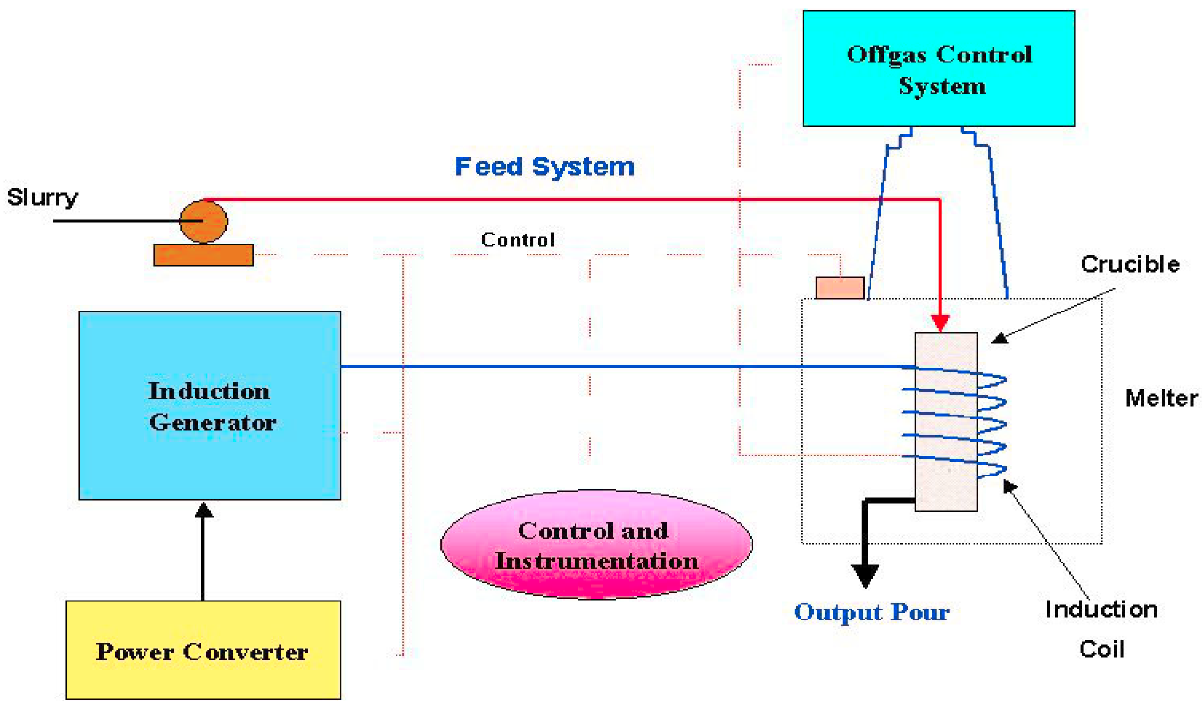

Figure 1 Simplified Schematic of the Major INL Bench Scale CCIM Systems

Operated by the Battelle Energy Alliance (BEA) since 2003, the CCIM is the only waste processing test melter that is of an induction cold-wall design in the United States. A unique design opposed to the traditional Joule-heated design. The latter design is being utilized at DOE's Savannah River Site (SRS) and will be deployed at DOE's Hanford site to vitrify defense liquid high-level-waste (HLW) into glass. However, past and present testing in CCIM pilot plants and international full-scale operations in both France and the Former Soviet Union (FSU) indicate some clear advantages of the CCIM over the conventional JHMs; all of which may prove beneficial for the calcine waste. These benefits are a direct result of the CCIM's unique design features (i.e., no electrodes or melter refractory exposed to corrosion at high temperatures) that provide for higher temperature melter operations compared to the JHM (i.e., up to 2000 degrees C compared to only 1000 degrees C). As a result, higher HLW loadings, greater waste throughputs and smaller footprints for the melter space are potentially achievable with the CCIM relative to the JHM. As such, if the CCIM/calcine test proves acceptable in the objectives outlined in the next section, the CCIM may be an acceptable cost, schedule, and technically risk adverse alternative to the base line calcine treatment option (i.e., HIPing).

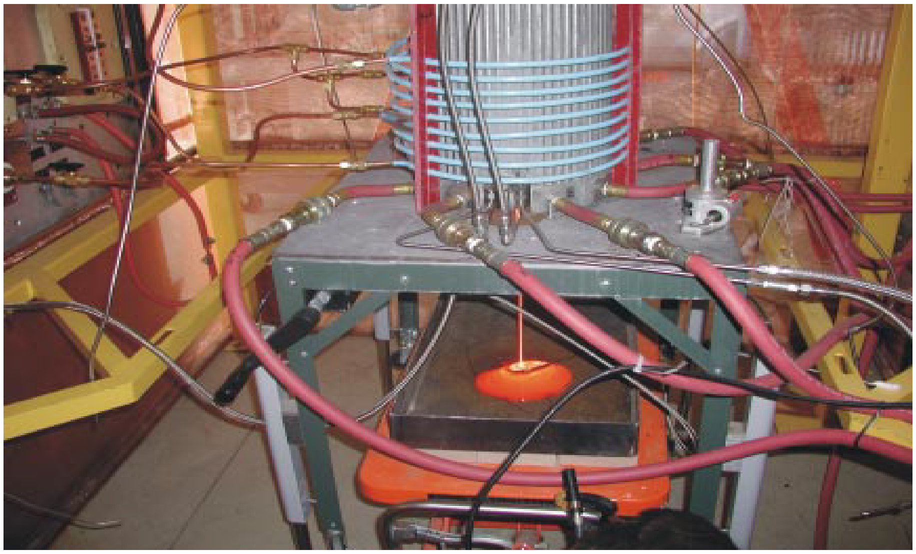

Figure 2 Photo of the INL CCIM Melter during a Melt Drain

3. Test Objectives

The first CCIM/calcine simulant test will be conducted to meet the following objectives based on discussions with CWI (the INL clean-up contractor) and DOE.

1. Stabilize the simulated HLW calcine into an amorphous vitrified form (as opposed to a glass-ceramic) at a mass waste loading as validated and recommended by the university pre-screening experiments (e.g., 35 -65 % waste loading is anticipated).
2. Determine the fate of key calcine simulant constituents – especially the partitioning of the Cd and Cr between the melt and the melter off-gas.
3. Maintain consistent operations for at least a 24-hour period to ensure steady state, semi –continuous operations and indicate proof of principle for use of the CCIM in vitrifying a solid particulate feed (e.g., the calcine).
4. Demonstrate dust-free, safe and efficient operation for the dry feed system to be installed above the bench-scale CCIM prior to testing.
5. Through post analyses determine the glass's acceptable or non-acceptable stabilization of the RCRA hazardous metals (i.e., mainly Cd and Cr-fed as oxides) spiked and incorporated in the surrogate calcine feed.
6. Through post analysis involving the standardized Product Consistency test (PCT) and/or the Vapor Hydration test (VHT) determine the glass's overall leachability and durability relative to other HLW glasses, especially to those consisting of a common borosilicate glass (BSG) matrix
7. Through a scaling calculation involving; 1) the ratio of the bench -scale CCIM's melt surface area and the surrogate calcine's (plus glass formulation additives and stabilizers) feed rate, 2) the corresponding nominal design and necessary feed rate of the actual calcine in an operational scale CCIM and 3) knowledge of the State of Idaho's calcine treatment milestone deadline, determine if a preliminary full-scale CCIM design has a foot-print suitable for the IWTU.

4. Chronological Test Activities

4.1 Contracted University Pre-screening Tests for Glass Formulations

Small scale multiple prescreening glass formulation tests are necessary prior to the actual scheduled testing in the INL's CCIM. The objective of these tests will be to identify the optimum glass formulation recipe for the surrogate calcine that takes advantage of the higher temperature features of the CCIM.

There are only a handful of University (and associated university affiliatives) that have the extensive experience and pedigreed expertise to conduct preliminary research and development involving the material science of vitrifying HLW into an efficient and durable waste form –suitable for eventual disposal. Based on this limited domestic capability, their excellent past work performance relationship with the INL, and their expertise in iron –phosphate-based glasses as opposed to the more traditional borosilicate glass; the INL has opted to sole source contract the University of Missouri Science and Technology (Rolla, Mo) and its associated industrial partner MoSci for the necessary preliminary calcine glass formulation tests. The test will be laboratory scale in nature and most likely involve small –scale parametric evaluations in crucibles. It is anticipated that one parametric may involve waste loadings (i.e., phase 1 testing) and another will involve melter temperature increases (i.e., phase 2 testing) under conditions of the general iron phosphate glass formulation as determined by MS&T and MoSci. The goal is that recommended formulations for the INL –CCIM/ test will eventually be those that achieve the highest waste loading that successfully passes performance testing (see section 4.4) at a temperature high enough to take advantage of the CCIM but also avoid unwanted component volatility to the off -gas

Once the defined surrogate calcine (refer to the section below on calcine surrogate formulation) is shipped to the university, the plan is to have them spike the calcine with the appropriate amounts of the appropriate heavy metals prior to instigating phase I of a planned 2 phase pre-screening evaluation. Specifics on the requirements and objectives of each test phase will be clearly defined in a Scope –of –Work (SOW)

4.2 Pre –Run Equipment Modifications

Prior to operating the CCIM for a sold feed surrogate waste condition, there are a number of critical modifications that must be completed as a minimum; they are as follows:

- Removal of the current CCIM liquid simulant feed system.
- Design, installation, and testing of a dry solid feed system suitable for dry feeding the calcine surrogate (as detailed in the below sub -section)
- Design and installation of dry augur feed system access and vents to ensure minimal and regulatory complaint worker exposure to the RCRA hazardous metal(s) in the dry particulate feed that constitutes the surrogate calcine.
- Resetting of the CCIM's radio frequency generator frequency to ensure inductive coupling of the generated magnetic field to the unique electrical resistivity of the surrogate calcine
- Revision of the approved Laboratory Instruction in a manner to accommodate for the hazards associated with a particulate feed containing RCRA hazardous metals
- Modification of the existing INL bench –scale CCIM air permit

4.3 Calcine Surrogate Formulation

The calcine simulant to be used in the first CCIM test run described in this test plan is very representative of the actual HLW aluminum nitrate/sodium blend calcine that is in INL bin set 6. The simulant to be used will be obtained from ~20,500 kgs of stored non -radioactive calcine that was formulated during the 1982 cold start-up of the (now shut down) INL New Waste Calcining Facility (NWCF). This cold calcine which is ~76%-64% aluminum oxide and 6.1%-1% dolomite was removed from the NWCF prior to NWCF "hot" operations and then stored in one of two temporary buildings. As such it has been given the designation of *T-2 calcine*.

In particular the T-2 is a mixture of startup bed dolomite (containing calcium/magnesium oxides and carbonates) and 3 different feeds. The three feeds were similar in that they were primarily aluminum nitrate, with varying amounts of sodium nitrate and some boron oxide. Since no RCRA metals were used in making the T-2 calcine, the mixture must be spiked with RCRA metals (i.e., at the least Cadmium [Cd] at a concentration of 55, 000 ppm [5.5 wt %]) to reflect the composition of the actual HLW calcine and ensure the objectives of the CCIM test run are achieved. Surrogate calcine compositions of cadmium will be achieved by spiking the T-2 calcine with amorphous cadmium oxide. It s is anticipated that ~ 25 kgs of spiked T-2 calcine will be required for the University tests and another 250 kgs of spiked T-2 calcine will be necessary for the INL bench –scale CCIM scheduled for February of 2012.

The entire existing radioactive HLW calcine stored in the 6 bin sets at the INL can be classified into three broad categories for CCIM test purposes; zirconium oxide based, aluminum oxide based, and a calcine consisting of an aluminum nitrate/sodium blend. The classification corresponds to the cladding metal of the original reprocessed fuel. In terms of actual calcine volume, about 75% of is zirconium based and the remaining 25% is about equal amounts of aluminum based and m the aluminum nitrate/sodium blend. As such, the T-2 calcine is representative of only12-13% of the total calcine inventory (i.e., the aluminum nitrate/sodium blend).

4.4 Test Conditions and Parameters

Target operating conditions are shown in Table 1. The specified melter feed rate, glass temperature, and other conditions, such as the extent of the cold –cap (if any) will be re-evaluated to determine if any changes should be considered based on a pre-run shake down test of the CCIM, university pre-screening evaluation results, and any applicable information received from DOE or the INL clean –up contractor after the issuance of this generalized test plan.

Table 1 Preliminary Target operating conditions for the 1st Calcine test in the INL CCIM.

Parameter	**Units**	**Test 1 – Baseline condition- no cold cap, baseline melt Temperature**	**Test 2 – Reduced cold cap, baseline melt Temperature**	**Test 3 – Higher melt Temperature, reduced cold cap**
Melter Feed: T-2 calcine with university recommended additives				
Melter feed rate	kg/hr	Highest rate (~2 kg/hr) with nearly no cold cap and a baseline melt	Highest rate (2-3kg/hr) with a minimal cold cap and a baseline melt	Highest rate(2-3kg/hr) with a minimal cold cap and a higher melt

		temp.	temperature	temperature
Duration	Hrs	≥48	≥24	>24
Melter:				
Induction power	kW	Sufficient for the target glass Temp.	Sufficient for the target glass Temp.	Sufficient for the target glass T
Cold cap coverage	%	~0-10%	~25%	~25%
Glass Temperature	^oC	1,250	1,250	1,500
Air bubbler rate	slpm	1	1	1
Freeboard gas Temperature	^oC	Typically 300-600^{o}C, with nearly no cold cap coverage	Typically 200-500^{o}C with partial cold cap coverage	Typically 400-700^{o}C, with partial cold cap coverage
Freeboard pressure	Inches of H_2O	0 to -0.5 inch	Same	Same
Off gas pipe T	^oC	Match freeboard gas Temperature	Same	Same
Scope of sampling/ measurements:				
Product glass rate, samples of product glass for PCT test and TCLP test		X	X	X
Off-gas flow rate and, composition (especially for Cr and Cd)		X	X	X
Off-gas PM and metals rate and composition		X	X	X
Total scrubber water volume and composition especially for Cr and Cd)		X	X	X
Notes: Slpm - Standard liters per minute. PM = Particulate matter.				

5. Final Report Schedule and FY-11 Milestones

The budget for the CCIM/Calcine test is divided into Fiscal years 2011 and 2012. The final report for all activities, including the CCIM test run results with T-2 calcine is tentatively set for May 21, 2012. Expenditure of approx. $195k in FY-2011 is required to meet the customer established FY-2011 deliverables which involve the following. The first two deliverables have milestone dates of September 30, 2011.

1. A status report on the identification, design, and partial installation of all equipment modifications required to ensure successful operations of the bench-scale CCIM with a simulated HLW calcine feed. Since this is the first time the bench-scale CCIM will receive feed of a solid nature containing RCRA hazardous materials, design and installation of a dust-free enclosure for the solid feeding system is necessary and will be a mandatory modification.

2. A status report on any progress made by the selected university with regard to the proposed pre-screening tests described earlier. As a minimum, the contract will be awarded, and the appropriate amount of RCRA metals to be spiked to the T2 calcine as well as an adequate amount of T2 calcine to complete the screening tests will be located and identified. In addition pre-screening test objectives will be clearly defined. The current budget allows for $120k for subcontracting the selected university. However, as little as $50k will be allocated in FY-11 to initiate phase 1 of 2 pre-screening test phases (as indicated by the schedule). This will provide sufficient FY-11 funds to complete all the milestones as well as ensure enough carry -over (CO) to maintain continuity given a continuing resolution CR in FY-12. BEA has approx. $213k available in unobligated funds in 2011 for this effort as of the end of July 20, 2011

Enough funds are available to complete another milestone indirectly connected to this test plan. The activity is related to an earlier FY-11 CCIM test run to validate the CCIM advantage in vitrifying a simulated liquid Hanford waste stream containing relativity high sulfur contents that are not appreciably contained in conventional (BSG). The milestone consists of completion of that test run's result that utilized an iron-phosphate glass. This glass type can immobilize a high waste loading of the sulfur-containing Hanford waste at the higher temperatures of the CCIM. The report's milestone due date is September 15, 2012.

6. References

1. Soelberg, Nick and Jay Roach, "Art CCIM Off-Gas System Evaluation Test Plan," INL-EXT-08-14449, January 2009.

2. D.Gombert, J.R.Richardson, "Cold Crucible Induction Melter Design and Development" Nuclear Technology 141 (2003) pp. 301-308